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#1. E. Krotscheck, G.-X. Qian, & W. Kohn, *Theory of inhomogeneous quantum systems. I. Static properties of Bose fluids* Physical Review B **31** 4245 (1985).

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Landauer Formula for the Current through an Interacting Electron Region

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A Landauer formula for the current through a region of interacting electrons is derived using the nonequilibrium Keldysh formalism. The case of proportionate coupling to the left and right leads, where the formula takes an especially simple form, is studied in more detail. Two particular examples where interactions give rise to novel effects in the current are discussed: In the Kondo regime, an enhanced conductance is predicted, while a suppressed conductance is predicted for tunneling through a quantum dot in the fractional quantum Hall regime.

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The formulation by Landauer [1] and Büttiker [2] of the current through a finite, possibly disordered region of noninteracting electrons has tremendously enhanced the understanding of transport in mesoscopic systems [3]. The Landauer formula, which expresses the current in terms of local properties of the finite region (such as the transmission coefficient) and the distribution functions in connected reservoirs, has been used extensively and successfully in many areas, including the scaling theory of localization [4,5], universal conductance fluctuations [6], Aharonov-Bohm conductance oscillations [7], the integer quantum Hall effect [8] and its quenching [9], the quantization of ballistic conductance [10], and recently in the field of quantum dynamics of driven systems (quantum chaos) [11].

While both the derivation of the Landauer formula for noninteracting electrons [3] and its application are well established, an apt formulation of the current when interactions between electrons are involved has been lacking. In view of the recent technological progress in confinement of electrons into small regions, where the electron-electron interactions plays a major role in the transport [12], it is quite clear that a Landauer-type formula for the transport through such an interacting region is highly desirable. Several attempts have been made to deal with special cases [13]. Langreth [14] was able to express the linear conductance through a single site with an on-site interaction (the Anderson model) at zero temperature in terms of phase shifts and thus relate the conductance to a scattering matrix. Apel and Rice [15] approximated the interaction in one dimension by the values of the momentum transfer δq at $\delta q = 0$ and $\delta q = 2k_f$ and were able to derive a Landauer-like formula. Unfortunately, this approximation is unsuitable for electrons confined into a small region. More recently, Hershfield, Davis, and Wilkins [16] have been able to derive a formula for the current in the Anderson model.

In this Letter (a) we derive an exact formula for the current through a region of interacting electrons coupled to two multichannel leads where the electrons are not interacting. The formula we derive [Eq. (6)] expresses the current, as in the noninteracting case, in terms of the Fermi functions in the leads and local properties of the interacting region. (b) We show how the noninteracting case and the results of Langreth and of Hershfield, Davis, and Wilkins follow as special cases. (c) The current will be written in a particularly simple form for the case of a constant asymmetry factor relating the coupling to the left lead to the coupling to the right lead. This case will be presented and investigated in some detail. (d) Two examples where the interactions lead to interesting results for the current will be discussed: transport in the Kondo regime, where an *enhanced* conductance has been predicted [17], and tunneling through a correlated electronic state, such as the fractional quantum Hall state, where a *suppression* of the conductance is expected [18].

Our starting point is the Hamiltonian

$$H = \sum_{k,\alpha \in L,R} \epsilon_{k\alpha} c^{\dagger}_{k\alpha} c_{k\alpha} + H_{int}(\{d^{\dagger}_n\};\{d_n\}) + \sum_{k,\alpha \in L,R} \sum_n (V_{k\alpha,n} c^{\dagger}_{k\alpha} d_n + \text{H.c.}), \quad (1)$$

where $c^{\dagger}_{k\alpha}$ ($c_{k\alpha}$) creates (destroys) an electron with momentum k in channel α in either the left (L) or the right (R) lead, and $\{d^{\dagger}_n\}$ and $\{d_n\}$ form a complete, orthonormal set of single-electron creation and annihilation operators in the interacting region. The channel index includes spin and all other quantum numbers which, in addition to k, are necessary to define uniquely a state in the leads. This Hamiltonian corresponds to an experimental situation (see, e.g., Refs. [10] and [12]) where two metallic, multichannel leads are connected to the mesoscopic system under study (see Fig. 1). Since the leads are metallic, the interaction between electrons in the leads and the interaction between electrons in the intermediate region and electrons in the leads are strongly screened and can be neglected. However, in the presence of barriers between the leads and the intermediate region, electrons cannot flow freely to screen the interactions in the intermediate region. Accordingly, the interactions between

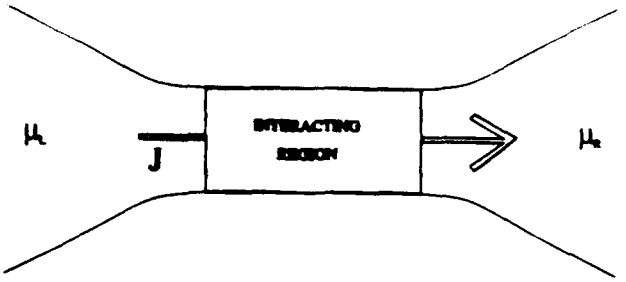

FIG. 1. Schematic diagram of the experimental configuration for which an *interacting* Landauer formula for the current is derived. Two leads, characterized by chemical potentials μ_L and μ_R, are connected to a mesoscopic region where electrons may interact. If $\mu_L > \mu_R$, an electron current J will flow from left to right.

electrons in the intermediate region need to be treated dynamically and are included in Eq. (1). This treatment is similar to the one leading to the Anderson model of a single magnetic impurity in a metal [19].

The logic of our approach follows that used in Ref. [20] for the one-dimensional noninteracting case. The unperturbed system (taken to exist at $t=-\infty$) consists of three uncoupled regions: a left lead and a right lead, both described by the first term in (1), and an interacting, intermediate region described by the second term in (1). Since the leads are not coupled at $t=-\infty$, each one maintains its own thermal equilibrium and one can associate chemical potentials, μ_L and μ_R, with the left and right leads, respectively. As the coupling turns on between the intermediate region and the two leads [the last term in (1)], then, if $\mu_L > \mu_R$, an electron current J starts to flow from the left lead to the right lead. After some time the system achieves a steady state. Our aim is to relate the steady state current to μ_L and μ_R, or equivalently, to $f_L(\epsilon)$ and $f_R(\epsilon)$, the unperturbed Fermi-Dirac distribution functions in the leads. (We assume that the reservoirs are large enough that the bulk μ_L and μ_R are not perturbed by the current J.)

To this end we write the current between the intermediate region and the left lead as

$$J=\frac{ie}{\hbar}\sum_{k,\alpha\in L,n}(V_{k\alpha,n}\langle c^{\dagger}_{k\alpha}d_n\rangle-V^{*}_{k\alpha,n}\langle d^{\dagger}_n c_{k\alpha}\rangle)$$
$$=\frac{e}{\hbar}\sum_{k,\alpha\in L,n}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}[V_{k\alpha,n}G^{<}_{n,k\alpha}(\omega)-V^{*}_{k\alpha,n}G^{<}_{k\alpha,n}(\omega)]. \tag{2}$$

The first line in (2) can be easily checked by writing the continuity equation for the current [20], while to get the second line we have used the definition of the Keldysh Green function [20,21], $G^{<}_{n,k\alpha}(t)\equiv i\langle c^{\dagger}_{k\alpha}d_n(t)\rangle$. In the Keldysh formalism, since the Hamiltonian describing the leads is noninteracting, one has the Dyson equations

$$G^{<}_{k\alpha,n}(\omega)=\sum_m V_{k\alpha,m}[g^{r}_{k\alpha,k\alpha}(\omega)G^{<}_{m,n}(\omega)-g^{<}_{k\alpha,k\alpha}(\omega)G^{a}_{m,n}(\omega)],$$
$$G^{<}_{n,k\alpha}(\omega)=\sum_m V^{*}_{k\alpha,m}[g^{<}_{k\alpha,k\alpha}(\omega)G^{a}_{n,m}(\omega)-g^{a}_{k\alpha,k\alpha}(\omega)G^{<}_{n,m}(\omega)], \tag{3}$$

where $G^{<}_{n,m}(t)\equiv i\langle d^{\dagger}_m d_n(t)\rangle$, and $G^{>}_{n,m}(t)$, to be used later, is equal to $-i\langle d_n(t)d^{\dagger}_m\rangle$. The Green functions with superscripts t and $\bar{t}$ are the time-ordered and the anti-time-ordered Green functions, respectively [22], and the Green functions denoted with small g are the unperturbed Green functions (i.e., in the uncoupled system). Using the equalities [22] $G^{>}(\omega)+G^{<}(\omega)=G^{r}(\omega)+G^{a}(\omega)$ and $G^{>}(\omega)-G^{<}(\omega)=G^{r}(\omega)-G^{a}(\omega)$, where G^{r} (G^{a}) are the usual retarded (advanced) Green functions, and the relations

$$g^{<}_{k\alpha,k\alpha}(\omega)=2\pi i f_L(\omega)\delta(\omega-\epsilon_{k\alpha}),$$
$$g^{>}_{k\alpha,k\alpha}(\omega)=-2\pi i[1-f_L(\omega)]\delta(\omega-\epsilon_{k\alpha}), \tag{4}$$

where $\alpha\in L$, we find

$$J=\frac{ie}{\hbar}\sum_{\alpha\in L,n,m}\int d\epsilon\,\rho_\alpha(\epsilon)V_{\alpha,n}(\epsilon)V^{*}_{\alpha,m}(\epsilon)\times\{f_L(\epsilon)[G^{r}_{n,m}(\epsilon)-G^{a}_{n,m}(\epsilon)]+G^{<}_{n,m}(\epsilon)\}, \tag{5}$$

where $\rho_\alpha(\epsilon)$ is the density of states in channel α and $V_{\alpha,n}(\epsilon)$ equals $V_{k\alpha,n}$ for $\epsilon=\epsilon_{k\alpha}$. An equivalent formula can be derived for the current between the intermediate region and the right lead. Since, in steady state, the current is uniform, one can symmetrize Eq. (5), and using matrix notation for the level indices in the interacting region, we find

$$J=\frac{ie}{2h}\int d\epsilon\{\mathrm{tr}\{[f_L(\epsilon)\Gamma^{L}-f_R(\epsilon)\Gamma^{R}](\mathbf{G}^{r}-\mathbf{G}^{a})\}+\mathrm{tr}\{(\Gamma^{L}-\Gamma^{R})\mathbf{G}^{<}\}), \tag{6}$$

where $\Gamma^{L}_{n,m}=2\pi\sum_{\alpha\in L}\rho_\alpha(\epsilon)V_{\alpha,n}(\epsilon)V^{*}_{\alpha,m}(\epsilon)$, with $\Gamma^{R}_{n,m}$ defined similarly. In equilibrium, $f_L(\epsilon)=f_R(\epsilon)=f_{eq}(\epsilon)$, one has $\mathbf{G}^{<}=-f_{eq}(\mathbf{G}^{r}-\mathbf{G}^{a})$ and the current vanishes.

Equation (6) is the central result of this work. It expresses the current through the interacting region in terms of the distribution functions in the leads and local properties of the intermediate region, such as the occupation and the density of states. (The local density of states is proportional to the diagonal part of $\mathbf{G}^{r}-\mathbf{G}^{a}$, while $\mathbf{G}^{<}$ is a product of the density of states and the occupation [23].) Note that these are to be calculated *in the presence* of the leads.

For the noninteracting case one can write down Dyson equations for the Green functions in the intermediate region [20], $\mathbf{G}^{<}=if_L(\epsilon)\mathbf{G}^{r}\Gamma^{L}\mathbf{G}^{a}+if_R(\epsilon)\mathbf{G}^{r}\Gamma^{R}\mathbf{G}^{a}$ and $\mathbf{G}^{r}-\mathbf{G}^{a}=-i\mathbf{G}^{r}(\Gamma^{L}+\Gamma^{R})\mathbf{G}^{a}$, which enable us to rewrite (6) as

$$J=\frac{e}{h}\int d\epsilon[f_L(\epsilon)-f_R(\epsilon)]\,\mathrm{tr}\{\mathbf{G}^{a}\Gamma^{R}\mathbf{G}^{r}\Gamma^{L}\}. \tag{7}$$

Since the transmission coefficient from left to right is given by $t_{\alpha,\alpha'} \equiv 2\pi \sum_{n,m} \rho_\alpha^{1/2} \rho_{\alpha'}^{1/2} V_{\alpha,n}^* G_{n,m}^r V_{\alpha',m}$, with $\alpha \in R$ and $\alpha' \in L$, Eq. (7) reduces to

$$J = \frac{e}{h} \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] \, \mathrm{tr}\{tt^+(\epsilon)\}, \tag{8}$$

which is the usual two-terminal Landauer formula for the noninteracting case [24].

The conductance formula, Eq. (6), takes an especially simple form [25] for the case that the couplings to the leads differ only by a constant factor, $\Gamma^L(\epsilon) = \lambda \Gamma^R(\epsilon)$,

$$J = \frac{ie}{h} \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] \, \mathrm{tr}\{\Gamma(G^r - G^a)\}$$

$$= -\frac{2e}{h} \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] \, \mathrm{Im}[\mathrm{tr}\{\Gamma G^r\}], \tag{9}$$

where $\Gamma \equiv \Gamma^L \Gamma^R / (\Gamma^L + \Gamma^R)$. While this innocent looking formula resembles the noninteracting one, it should be emphasized that even though there is a single integral over energy in (9), this formula includes, by means of the full Green function G^r, inelastic processes, spin flips, and even processes where several electrons are scattered.

In order to illustrate how the additional processes due to interactions are reflected in the formula for the current, we use the usual definition of the self-energy Σ^r, namely, $\Sigma^r = (g^r)^{-1} - (G^r)^{-1}$, with Σ^a defined similarly, to write $G^r - G^a = G^r \Sigma G^a$, where $\Sigma = \Sigma^r - \Sigma^a$. This allows us to rewrite Eq. (9) as

$$J = \frac{ie}{h} \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] \, \mathrm{tr}\{\Gamma G^r \Sigma G^a\}$$

$$= \frac{e}{h} \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] \, \mathrm{tr}\{G^a \Gamma^R G \Gamma^L \Sigma_0^{-1} \Sigma\}, \tag{10}$$

where Σ_0, the self-energy for the noninteracting case, is equal to $-i(\Gamma^L + \Gamma^R)$. Comparing Eq. (10) to the noninteracting results, Eqs. (7) and (8), we see that in the presence of interactions the current cannot in general be recast in terms of the transmission matrix, and that the additional processes included in Σ are directly responsible for the deviation of the exact formula for the interacting case from the usual Landauer formula [Eq. (8)]. Note, however, that at zero temperature and in linear response only single-electron, elastic processes are allowed by energy conservation. In this case $\Sigma(\mu) = \Sigma_0(\mu)$ and Eq. (10) reduces to the usual Landauer formula (8) (this is a generalization of the Langreth [14] result for the Anderson model). Thus the Landauer formula for the linear-response conductance holds not only for the noninteracting case, but also for the interacting case at zero temperature. At finite temperature, or at finite voltage, inelastic processes have to be taken into account and the usual Landauer formula (8) breaks down for an interacting system.

In order to demonstrate the new features of Eq. (9), we now study two specific examples. The first is transport through a quantum dot in the Coulomb blockade regime [12]. Recently, the Anderson Hamiltonian [19],

$$H = \sum_{k,\sigma} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \epsilon_0 \sum_\sigma d_\sigma^\dagger d_\sigma + U n_\uparrow n_\downarrow + \sum_{k \in L,R; \sigma} (V_{k\sigma} c_{k\sigma}^\dagger d_\sigma + \mathrm{H.c.}), \tag{11}$$

has been employed [26] to describe transport in this regime. Equation (11) is a special case of (1), where the different channels are the two spin directions, and the interacting region is a single site with an on-site Coulomb repulsion U. In this case the current takes the form

$$J = \frac{e}{h} \sum_\sigma \int d\epsilon [f_L(\epsilon) - f_R(\epsilon)] \Gamma_{\sigma\sigma}(\epsilon) \left[-\frac{1}{\pi} \mathrm{Im} G_{\sigma\sigma}^r(\epsilon) \right], \tag{12}$$

where $-(1/\pi)\mathrm{Im}G_{\sigma\sigma}^r(\epsilon)$ is just the local density of states of electrons with spin σ. In linear response, for temperatures larger than the Kondo temperature, the conductance will consequently exhibit resonant tunneling peaks only at ϵ_0 and $\epsilon_0 + U$, which correspond to resonances in the density of states of the uncoupled site [26]. However, below the Kondo temperature, the density of states develops a peak at the Fermi energy [27], for $\epsilon_0 < \mu < \epsilon_0 + U$. Consequently, Eq. (12) predicts a greatly enhanced conductance over this entire range, in agreement with earlier studies [17]. Equation (12), which has been independently derived by Hershfield, Davis, and Wilkins [16] for constant Γ, provides a framework to study the crossover from high to low temperatures, and to calculate the current in a Kondo system out of equilibrium. Detailed studies will be presented elsewhere [28].

A second example where nontrivial effects due to interactions appear in the conductance is the case of tunneling through a quantum dot in a highly correlated state, such as a fractional quantum Hall state. In the case where the coupling to the leads is weak, i.e., the elastic broadening of the levels is smaller than the excitation energy, the correlated eigenstates are only weakly perturbed by the leads. When the temperature is larger than the elastic broadening one can rewrite Eq. (9) for the linear-response conductance G in the form

$$G = \frac{e^2}{h} \sum_{m,n} \Gamma_{n,m}(E_j - E_i) \sum_{i,j} (P_i + P_j) \left[-\frac{\partial f_{eq}}{\partial \epsilon}(E_j - E_i) \right] \langle \psi_j | d_n^\dagger | \psi_i \rangle \langle \psi_i | d_m | \psi_j \rangle, \tag{13}$$

where the ψ_i are eigenstates, with energies E_i, of the uncoupled interacting region, and P_i is the equilibrium probability of state ψ_i. Hence, the conductance consists of thermally broadened resonant tunneling peaks which occur whenever the chemical potential μ suffices to add another electron to the interacting region. For noninteracting electrons, one can choose the d's to correspond to single-particle eigenstates of the uncoupled system, and the overlap factor in each term, $\langle \psi_j | d_n^\dagger | \psi_i \rangle \langle \psi_i | d_m | \psi_j \rangle$, is trivially 0 or 1. On the other hand, adding an electron in a single-particle state to a correlated N-particle eigenstate—which cannot be written as a Slater determinant of single-particle states—will generally not pro-

duce an $(N+1)$-particle eigenstate, and consequently these overlap factors can reduce the conductance significantly. In particular, the conductance through a quantum dot in the fractional quantum Hall regime has been studied in detail in Ref. [18] where it was shown that the overlap factors reduce the conductance peaks by a factor of $1/N^{(p-1)/2}$, for the $\nu=1/p$ Laughlin state, where N is the number of electrons. Thus Eq. (13) predicts a strong suppression of the conductance in this regime, an effect of considerable experimental significance. Moreover, formula (13) provides a way to calculate the conductance in more complicated fractional quantum Hall states (such as $\nu=\frac{2}{5}$), where even more interesting effects, such as an alternating suppression of the conductance peaks, are predicted to occur [18].

To conclude, we have derived a Landauer-type formula for the current through an interacting electron region. This formula provides a new framework to study transport in mesoscopic systems where interaction effects are important. We have demonstrated the usefulness of this new tool for interacting systems by identifying novel features in the conductance for two examples: the Kondo regime and the fractional quantum Hall regime. Further investigations are currently under way.

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$$J=\frac{ie}{2h}\int d\epsilon\{\mathrm{tr}[\Gamma^{L}[f_{L}(\epsilon)-F](G^{r}-G^{a})] + \mathrm{tr}[\Gamma^{R}[F-f_{R}(\epsilon)](G^{r}-G^{a})]\},$$

which shows that the current can be expressed in terms of a difference in distribution functions times the local density of states. Note, however, that in order to calculate the local density of states and the local distribution matrix one has to calculate the Green functions G^{r}, G^{a}, and $G^{<}$.

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Surface Effects on Bulk Plasmons

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We discuss the effects of surfaces on bulk plasma oscillations in a metallic film of thickness L. The leading corrections to the plasma frequencies due to the surfaces are proportional to L^{-1}. The frequencies of low-lying long wavelength modes are given by the dispersion relation of bulk modes, $\omega = \omega_p[1 + \alpha(q^2/k_F^2)]$, where the allowed q-values are $q^{(n)} = (n\pi/\overline{L})$, $\overline{L} \equiv L - 2d_b$, and d_b is a complex length characteristic of the surface (reminiscent of but different from Feibelman's $d_\perp$). An explicit expression for d_b is derived. Resonant excitation by an external field, $E_0 e^{-i\omega t}$, is calculated.

The widespread interest in nanostructures (thin films, layer structures, quantum wires, quantum dots *etc.*) as well as in atomic and molecular clusters, in which at least one dimension is "mesoscopic" (10 Å - 1000 Å), has led us to re-examine the effects of finite size on charge collective modes (plasmons). In an infinite system, these modes have frequencies given by the bulk plasmon relation [1]

$$\omega(q) = \omega_p + \alpha \left(\frac{q}{k_F}\right)^2 \omega_p, \tag{1}$$

where ω_p is the classical plasma frequency, $\omega_p^2 = 4\pi e^2 n/m$, with n the electron density, k_F the Fermi wavevector, and α a (complex) number. The imaginary part of α describes the damping of bulk plasmons, which in an infinite system is due to excitations of multiple electron-hole pairs. In a finite system, we expect the following qualitative changes in the plasmon spectrum due to the presence of surfaces. First, only a discrete set of frequencies will be allowed. Second, there will be additional damping due to the decay of plasmons into (single) electron-hole pairs. In this Letter, we will show how both the real and imaginary parts of the allowed frequencies are completely determined by a single (complex) length. Specifically, we consider a thin slab with a positive jellium charge background, $n_+ = \overline{n}$ between $z = 0$ and $z = L$ and infinite in the x and y directions, and a neutralizing electron liquid described by the equilibrium density distribution $n_-(z)$ (See Fig. 1). We shall discuss bulk-like collective modes of the form

$$n(\mathbf{r}, t) = n(z) e^{-i\omega^{(n)} t}, \tag{2}$$

where the eigenfrequencies $\omega^{(n)}$ are near the classical bulk plasma frequency associated with the density $\overline{n}$, $\omega_p = (4\pi e^2 \overline{n}/m)^{1/2}$.

This problem has been previously addressed experimentally [2,3] and theoretically [4] for modes of the form (2) and of the more general form

$$n(\mathbf{r}, t) = n(z) e^{i(px - \omega t)}. \tag{3}$$

In the present Letter, limited to the case $p = 0$, we derive the following new result: the (complex) eigenfrequencies $\omega^{(n)}$, near ω_p, are determined by the bulk plasmon dispersion Eq. (1) for small q, and by an effective, complex, slab thickness

$$\overline{L} = L - 2d_b, \tag{4}$$

where d_b, precisely defined below, is an effective, complex, surface thickness. The thickness d_b is reminiscent of the parameter $d_\perp$ introduced by Lang and Kohn [5] for $\omega = 0$ and generalized for arbitrary ω by Feibelman [6] but is *not* the same quantity. The allowed (complex) interior wavenumbers, $q^{(n)}$, are given by

$$q^{(n)} \overline{L} = n\pi \quad (n = 1, 2, \ldots) \tag{5}$$

and the corresponding (complex) eigenfrequencies are given by Eq. (1):

$$\omega^{(n)} = \omega(q^{(n)}). \tag{6}$$

We now derive the above-stated results. We take the equilibrium electron density, $n_-(z)$, to vanish for $z < -c$ and $z > L + c$, and introduce an auxiliary thickness [7] a, large compared to the surface thickness but much smaller than L. We then distinguish two surface regions, $-c < z < a$ and $L - a < z < L + c$, and the bulk region $a < z < L - a$, in which $n_-(z) = \overline{n}$ (see Fig. 1).

We consider a collective mode of frequency ω, with self-consistent electric field, $E(z)$, parallel to z. This field induces a current density,

$$j(z) = \int_{-c}^{L+c} dz' \sigma(z, z') E(z'), \tag{7}$$

where $\sigma(z, z') = \int dx' dy' \sigma_{zz}(x - x', y - y'; z, z')$; σ_{zz} is the zz-component of the non-local conductivity tensor. (For ease of notation, the time dependence, $e^{-i\omega t}$, and the functional dependence of fields, currents *etc.* on ω will generally not be explicitly written out.) The quantity $\sigma(z, z')$ is short-ranged in the difference variable $(z - z')$; also, when both z and z' are in the bulk region,

$$\sigma(z, z') = \sigma(z - z'), \quad a < z, z' < L - a \tag{8}$$

equal to the bulk conductivity corresponding to $\overline{n}$ and ω. Of course $\sigma(z, z')$ vanishes when either z or z' are outside the slab $(-c, L + c)$, as do $E(z)$ (due to charge neutrality) and $j(z)$.

Denoting the induced charge density by $n(z)$, the continuity equation and Gauss' law are

$$i\omega e n(z) + \frac{dj(z)}{dz} = 0 \tag{9}$$

and

$$\frac{dE(z)}{dz} = -4\pi e n(z) \tag{10}$$

(where e has been taken as positive). Since at $z = -c$ both $j(z)$ and $E(z)$ vanish, (9) and (10) imply that, for *all* z,

$$j(z) = \frac{i\omega}{4\pi} E(z). \tag{11}$$

We can eliminate $j(z)$ from (7) and (11) which results in the integral equation

$$\frac{i\omega}{4\pi} E(z) = \int_{-c}^{L+c} dz' \sigma(z, z') E(z'). \tag{12}$$

We are looking for solutions, which, in the bulk, behave as $e^{\pm iqz}$, with $q \ll k_F$. In this limit of long wavelengths in the bulk, we can, with Eq. (8) in Eq. (12) obtain the frequencies of these solutions by expanding $E(z')$ about z when z and z' are in the bulk. The result is

$$\frac{i\omega}{4\pi} = \sigma_0(\omega) - \frac{1}{2}\sigma_2(\omega)q^2, \tag{13}$$

where

$$\sigma_0(\omega) = \int dz'\,\sigma(z-z';\omega) = \frac{i\omega_p^2}{4\pi\omega}, \tag{14}$$

$$\sigma_2(\omega) = \int dz'(z'-z)^2\sigma(z-z';\omega). \tag{15}$$

Equation (13) is the bulk plasmon dispersion relation of an infinite system of conductivity $\sigma(z-z')$. In the limit $L \gg a$, the frequencies of the lowest-lying eigenmodes of Eq. (12) are very near ω_p and the shape of $E(z)$ in the surface region tends to the limiting shape

$$E(z) \to \tilde{E}(z) \equiv E(z;\omega_p) \tag{16}$$

shown schematically in Fig. 2. We arbitrarily take $E(a) = 1$ and write, near $z = a$

$$\tilde{E}(z) = \frac{z - d_b}{a - d_b} \tag{17}$$

where d_b is the, as yet undetermined, point where $\tilde{E}(z)$ extrapolates to 0. When z is in the bulk the solution which joins (17) near $z = a$ is

$$E(z) = \frac{1}{q(a-d_b)}\sin q(z-d_b). \tag{18}$$

Thus, d_b plays the role of a (complex) scattering length.

The length d_b can be simply expressed in terms of the limiting surface density distribution, $\tilde{n}(z)$, associated with collective modes with $q \to 0$ and $\omega \to \omega_p$. From Eq. (10) and the form of $E(z)$ in the surface region we see that $\tilde{n}(z)$ has the general form shown in Fig. 2. Thus, for z near a, we have

$$\begin{aligned}\frac{\tilde{E}(z)}{(-4\pi e)} &= \int_{-c}^{z} dz'\,\tilde{n}(z) = \int_0^z dz'\left[\frac{d}{dz'}(z'\tilde{n}(z')) - z'\frac{d\tilde{n}(z')}{dz'}\right] \\ &= zn_{-c} - \int_{-c}^{\infty} dz'\,z'\frac{d\tilde{n}(z')}{dz'} \\ &= z\int_{-c}^{\infty} dz'\,\frac{d\tilde{n}(z')}{dz'} - \int_{-c}^{\infty} dz'\,z'\frac{d\tilde{n}(z')}{dz'}.\end{aligned} \tag{19}$$

The upper limits ∞ are to be understood as $\gg k_F^{-1}$ but $\ll L$. The linear form of $E(z)$ near a, Eq. (19), extrapolates to zero when z equals

$$d_b \equiv \frac{\int_{-c}^{\infty} dz\, z\frac{d\tilde{n}(z)}{dz}}{\int_{-c}^{\infty} dz\,\frac{d\tilde{n}(z)}{dz}}. \tag{20}$$

Thus, d_b is the center of mass of the function $d\tilde{n}(z)/dz$.

The eigenmodes are either even or odd about $L/2$ so that, by Eq. (18), the allowed values of q are

$$q^{(n)}\overline{L} = n\pi, \quad (n = 1, 2, \ldots) \tag{21}$$

where $\overline{L}$ is give by Eq. (4). The corresponding frequencies are given by Eq. (1). Using the notation $d_b = d_b' + id_b''$ *etc*, we have

$$\omega^{(n)\prime} = \omega_p + \frac{1}{k_F^2}\left(\alpha' - 4\alpha''\frac{d_b''}{\overline{L}'}\right)\left(\frac{n\pi}{\overline{L}'}\right)^2 \tag{22}$$

$$\omega^{(n)\prime\prime} = \frac{1}{k_F^2}\left(\alpha'' + 4\alpha'\frac{d_b''}{\overline{L}'}\right)\left(\frac{n\pi}{\overline{L}'}\right)^2, \tag{23}$$

where $\overline{L}' = L - 2d_b'$. We observe that the real parts Eq. (22) of the frequencies correspond to bulk plasmon modes with the boundary conditions $E(d_b') = E(L - d_b') = 0$. The imaginary parts, (23), have contributions from the bulk ($\propto \alpha''$) and from the surface ($\propto (d_b''/\overline{L}')$).

We have solved the integral equation (12) in the discrete form

$$\frac{i\omega}{4\pi}E_m = b\sum_{n=0}^{N}\sigma_{mn}E_n, \tag{24}$$

where $b \equiv L/N$ is the interval length. We have chosen $L = 35.4$ Å, $N = 248$, $b = 0.14$ Å, and $c = 0$. For the sake of illustration, we have chosen the following values for σ_{mn}. In the "bulk", $4 \leq m \leq 125$, we have chosen

$$\sigma_{m,m} = i\mu(\omega) = \frac{i\omega_p^2}{4\pi\omega b} - \frac{\omega_p}{3\pi k_F^2 b^3}\left[\alpha_2 - i\alpha_1\right], \quad \sigma_{m,m+2} = \sigma_{m,m-2} = \frac{1}{2}i\nu$$

$$\sigma_{m,m+1} = \sigma_{m+1,m} = i\nu = \frac{2\omega_p}{21\pi k_F^2 b^3}\left[\alpha_2 - i\alpha_1\right], \quad \sigma_{m,m+3} = \sigma_{m,m-3} = \frac{1}{4}i\nu \tag{25}$$

and symmetrically for $m > 125$, with $k_F = 1.75 \times 10^8$ cm^{-1}, $\hbar\omega_p = 15$ eV, and $\alpha = 0.57 + i0.025$ in Eq. (1) (appropriate [1] for Al in the limit of $q/k_F \ll 1$). The functions $\mu(\omega)$ and ν are chosen so that the average and second moment of σ_{mn} are $\sigma_0(\omega)$ and $\sigma_2(\omega_p)$. In the "surface", $0 \leq m \leq 3$, we have chosen

$$\begin{aligned}
\sigma_{0n} &= \sigma_{n0} = 0 \\
\sigma_{m,m} &= \frac{m}{4}\left(i\mu(\omega) + \epsilon\right) \\
\sigma_{m,m+1} &= \sigma_{m+1,m} = \frac{m}{4}\left(i\nu + \epsilon\right) \\
\sigma_{m,m+2} &= \sigma_{m+2,m} = \frac{m}{8}\left(i\nu + \epsilon\right) \\
\sigma_{m,m+3} &= \sigma_{m+3,m} = \frac{m}{16}\left(i\nu + \epsilon\right)
\end{aligned} \tag{26}$$

and symmetrically near the other surfaces. The small real part $\epsilon = \omega_p \alpha''/(6\pi k_F^3 b^3)$ was added to the conductivity in the surface regions as a simple model of damping in the surfaces due to decay into electron-hole pairs. Apart from this small real part, equation (26) is a simple interpolation between $\sigma_{0n} = \sigma_{n0} = 0$ and the bulk conductivity. The results for the lowest three modes are plotted in Figs. 3 and 4, and confirm our conclusions, Eqs. (5), (6), (22) and (23).

How do our results relate to charge density waves in classical Maxwell theory? "Classical" here means a local $\sigma(z,z')$

$$\sigma_{\text{class}}(z,z') = \sigma_0 \delta(z-z') = \frac{i\omega_p^2}{4\pi\omega}\delta(z-z'), \tag{27}$$

so that (12) becomes

$$\frac{i\omega}{4\pi}E(z) = \frac{i\omega_p^2}{4\pi\omega}E(z). \tag{28}$$

The boundary conditions [8] are, from charge neutrality, $E(0) = E(L) = 0$. Thus, classically the general solution is $\omega = \omega_p$ and $E(z) = F(z)$, an *arbitrary* function of z satisfying the boundary conditions $E(0) = E(L) = 0$. In particular, the functions

$$F_n(z) = \sin(q_{\text{class}}^{(n)} z) \quad q_{\text{class}}^{(n)} \equiv \frac{n\pi}{L},\ n = 0, 1, \ldots \tag{29}$$

$$\tilde{\omega}^{(n)} \equiv \omega_p, \quad \text{all } n, \tag{30}$$

are solutions. Thus, our *non-local* and translationally *non-invariant* $\sigma(z,z')$ leads to: (1) replacement of L by the complex $\overline{L} = L - 2d_b$; (2) a *finite* dispersion of $\omega^{(n)}$; and, if $\sigma(z,z')$ is known in detail in the surface region, the detailed field and density distributions in the surface region.

We now turn to resonant response to a uniform external field, $E_0 e^{-i\omega t}$. Eqs. (7), (9) and (10) remain unchanged, but in Eq. (11) $E(z)$ is replaced by $E(z) - E_0$ and Eq. (12) becomes

$$\frac{i\omega}{4\pi}[E(z) - E_0] = \int_{-c}^{L+c} dz'\, \sigma(z,z';\omega)E(z'). \tag{31}$$

Near $\omega = \omega_p$ we can write this as

$$\left[\frac{\omega_p}{4\pi}E(z) - \int dz' \frac{\sigma(z,z')}{i}E(z')\right] + \eta\left[\frac{1}{4\pi}E(z) - \int dz' \frac{\tau(z,z')}{i}E(z')\right] = \frac{(\omega_p+\eta)}{4\pi}E_0, \tag{32}$$

where $\sigma(z,z') \equiv \sigma(z,z';\omega_p)$, $\tau(z,z') \equiv [d\sigma(z,z';\omega)/d\omega]_{\omega=\omega_p}$ and $\eta \equiv \omega - \omega_p$. On the right-hand side, η can be neglected.

Setting $E_0 = 0$, for a moment, gives us the equation for the slightly complex normal modes, $E_n(z)$, and eigenvalues, $\eta_n = \eta_n' + i\eta_n''$. The kernels $\sigma(z,z')/i$, $\tau(z,z')/i$ are symmetric and (slightly) complex. The $E_n(z)$ satisfy the orthonormality relations

$$\int_{-c}^{L+c}\int_{-c}^{L+c} dz\,dz'\,E_m(z)\left[\frac{1}{4\pi}\delta(z-z')-\frac{\tau(z,z')}{i}\right]E_n(z')=\delta_{mn}. \tag{33}$$

We now substitute the Ansatz

$$E(z)=\sum_n A_n E_n(z) \tag{34}$$

into (32), giving

$$\sum_n A_n(\eta-\eta_n)E_n(z)=\frac{\omega_p}{4\pi}E_0. \tag{35}$$

Using the orthonormality relation Eq. (33) leads to

$$A_n=\frac{\omega_p E_0}{4\pi(\eta-\eta_n)}\int_{-c}^{L+c}dz\int_{-c}^{L+c}dz' E_n(z')\left[\frac{1}{4\pi}\delta(z-z')-\frac{\tau(z,z')}{i}\right]. \tag{36}$$

Only even modes, $n=2,4\ldots$ are excited. A well defined resonance occurs for $\eta\approx\eta'_n$ if $\eta''_n \ll |\eta'_n-\eta'_{n\pm2}|$. In this case as single term in (34) is dominant, giving

$$E_n(z)=E_0\frac{\omega_p\left[(\eta-\eta'_n)-i\eta''_n\right]}{4\pi\left[(\eta-\eta'_n)^2+\eta''^2_n\right]}. \tag{37}$$

In order to resolve the discrete modes experimentally, the spacing between the modes must be larger than the width. From Eqs. (22) and (23) we find that the spacing and width are given by

$$\left(\eta'_n-\eta'_{n-2}\right)=\frac{\alpha'}{k_F^2}\left(\frac{\pi}{L'}\right)^2(4n-4) \tag{38}$$

$$\eta''_n=\frac{1}{k_F^2}\left(\alpha''+\frac{4\alpha' d''_b}{L'}\right)\left(\frac{\pi}{L'}\right)^2 n^2. \tag{39}$$

Thus, as n increases, the width gains on the spacing. For a film of width $L'=30$ Å, $d'_b=d''_b=1$ Å, and $\alpha'=0.57$, $\alpha''=0.025$ (Al), the highest well defined resonance is $n\approx 20$.

We plan to address the considerably more complex theory of modes with finite wavevector p (Eq. 3) in the near future.

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FIGURES

FIG. 1. Positive background density, $n_+(z)$ (dashed line), and equilibrium electron density, $n_-(z)$ (dotted line).

FIG. 2. The limiting ($\omega \to \omega_p$) form, $\tilde{E}(z)$, and mode density $\tilde{n}(z)$ in the surface regions. The mode density $\tilde{n}(z)$ approaches the constant value n_0 in the bulk.

FIG. 3. (a) The real part of the electric field of the three lowest bulk-like eigenmodes near the surface. In the surface, the fields all approach the limiting form $\tilde{E}(z)$. *Insert* The real parts of the three lowest bulk-like eigenmodes for $0 < z < L/2$. In the bulk region, the fields are sinusoidal. The vertical dashed line marks the position of a.

FIG. 4. (a) Real and (b) imaginary parts of the plasmon eigenfrequencies. The straight lines are the dispersions Eqs. (22) and (23) with $L' = 34.8$ Å and $d_b'' = 6 \times 10^{-2}$ Å obtained from Eq. (20).

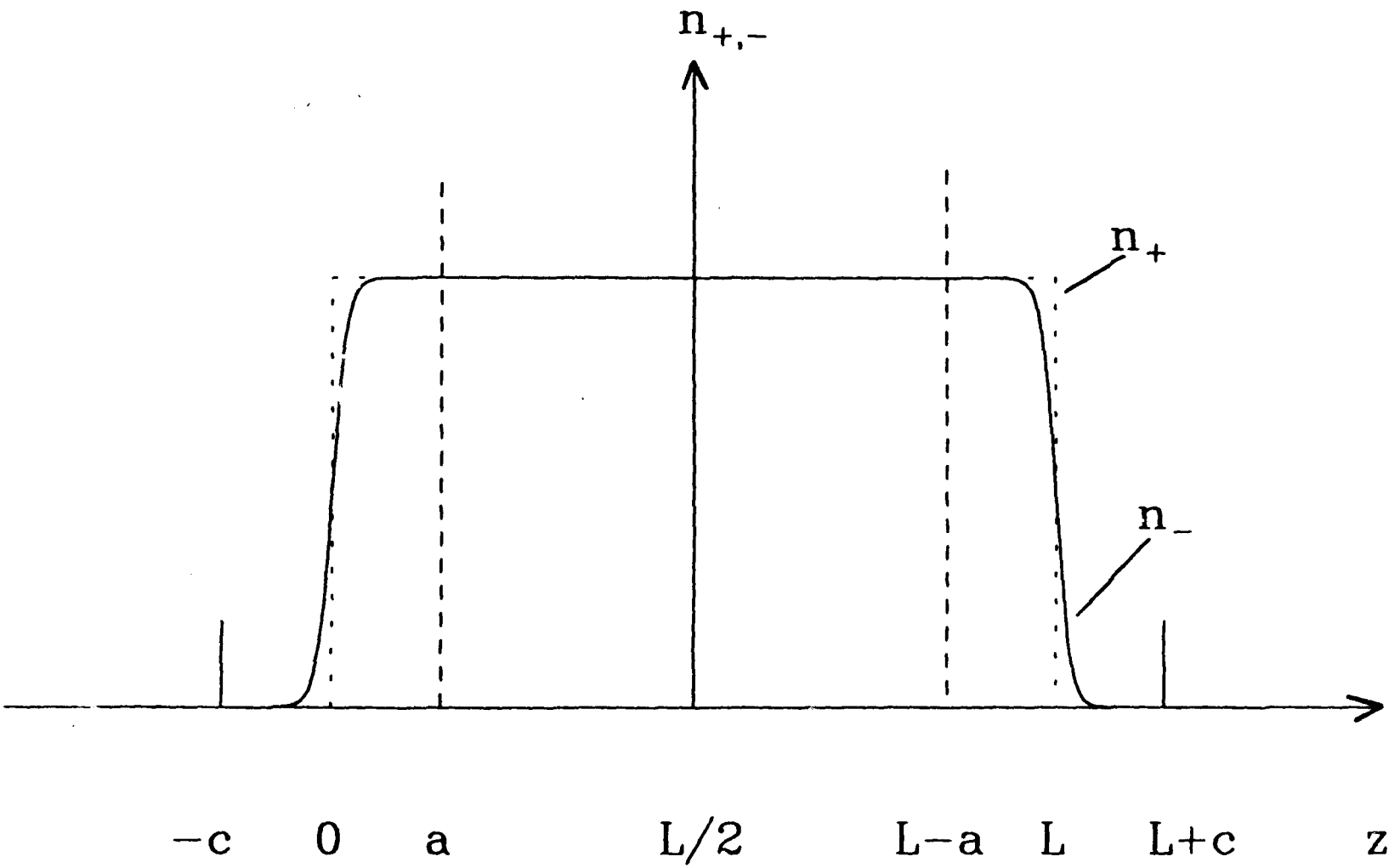

Fig 1

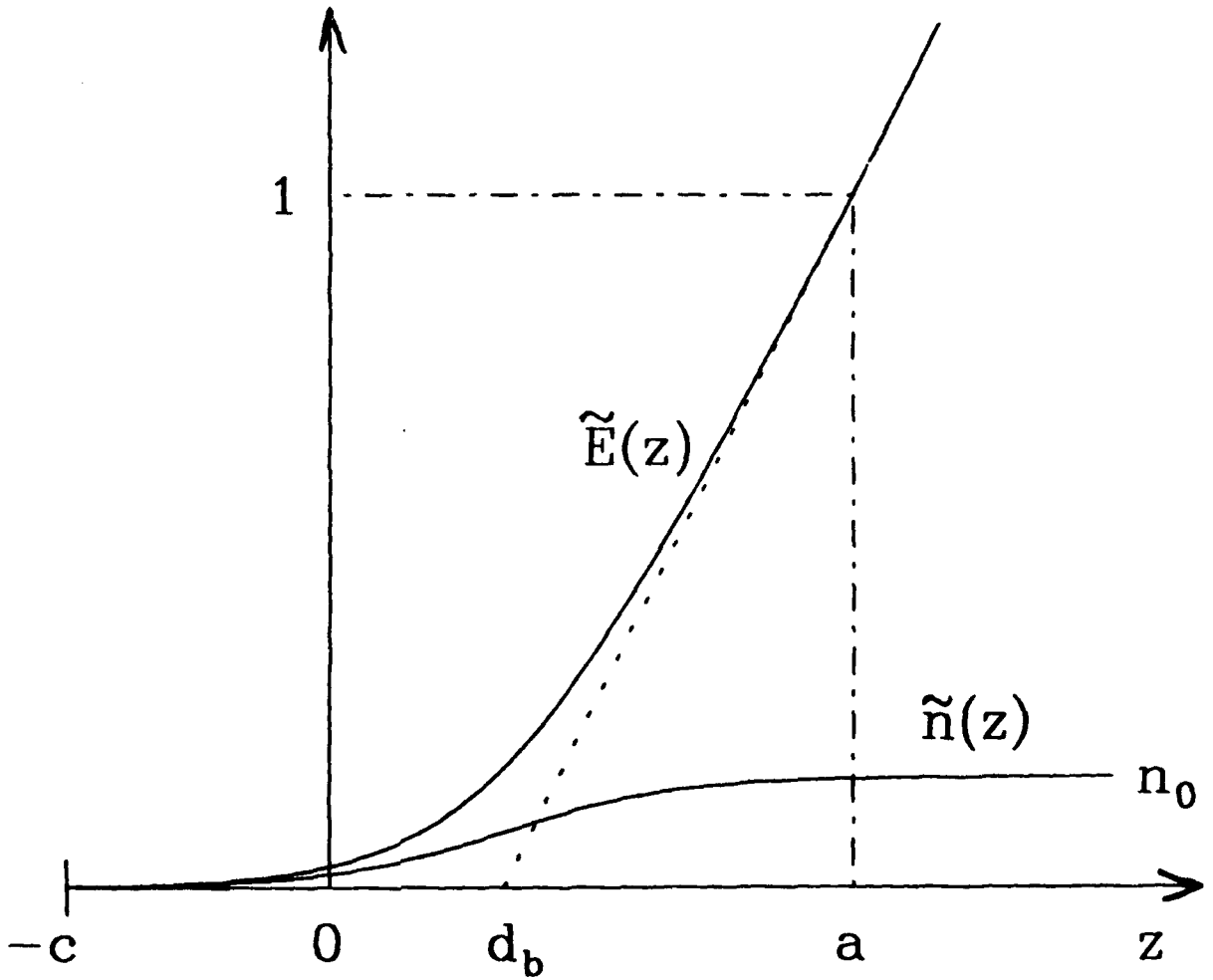

Fig. 2

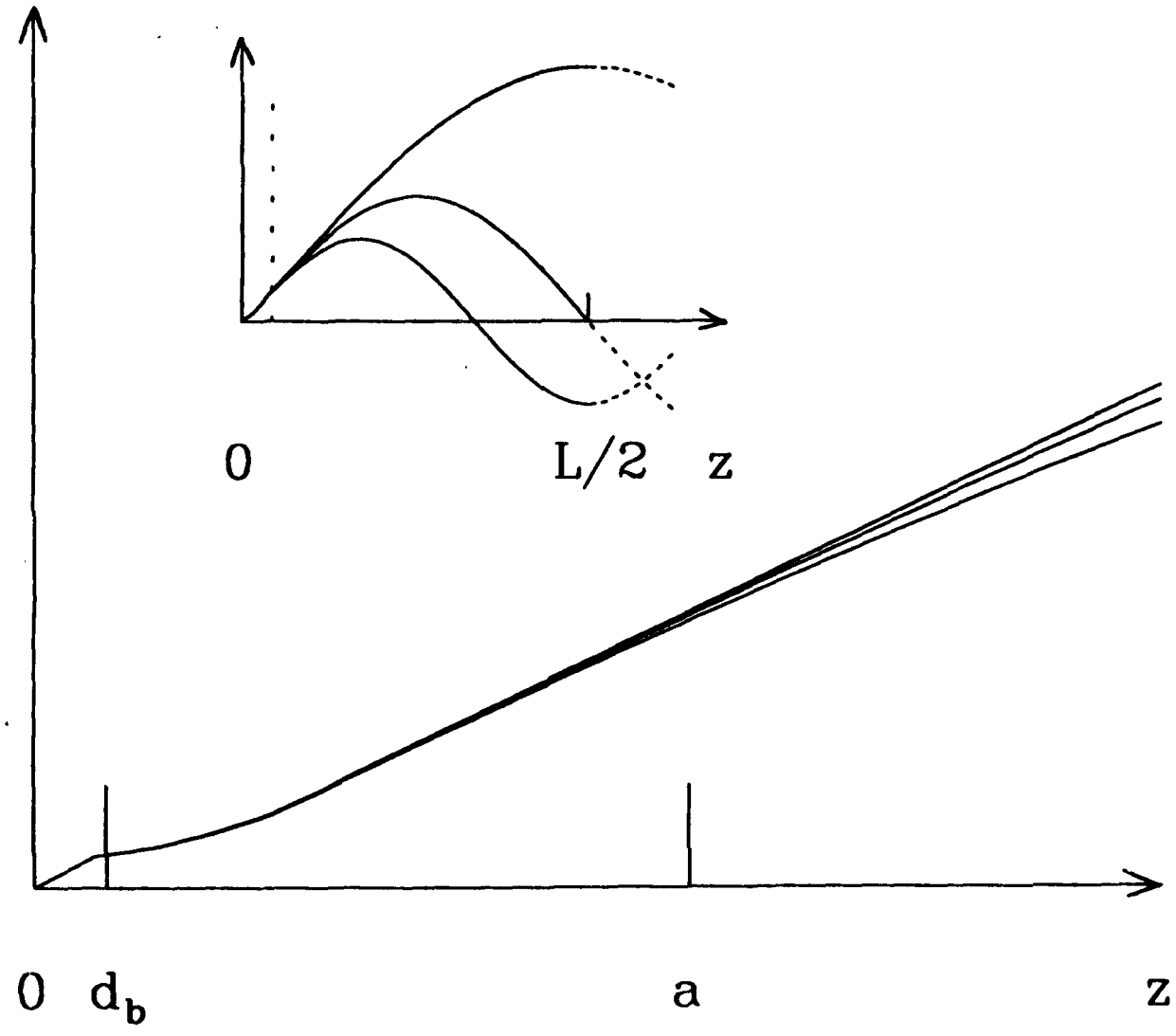

Fig 3

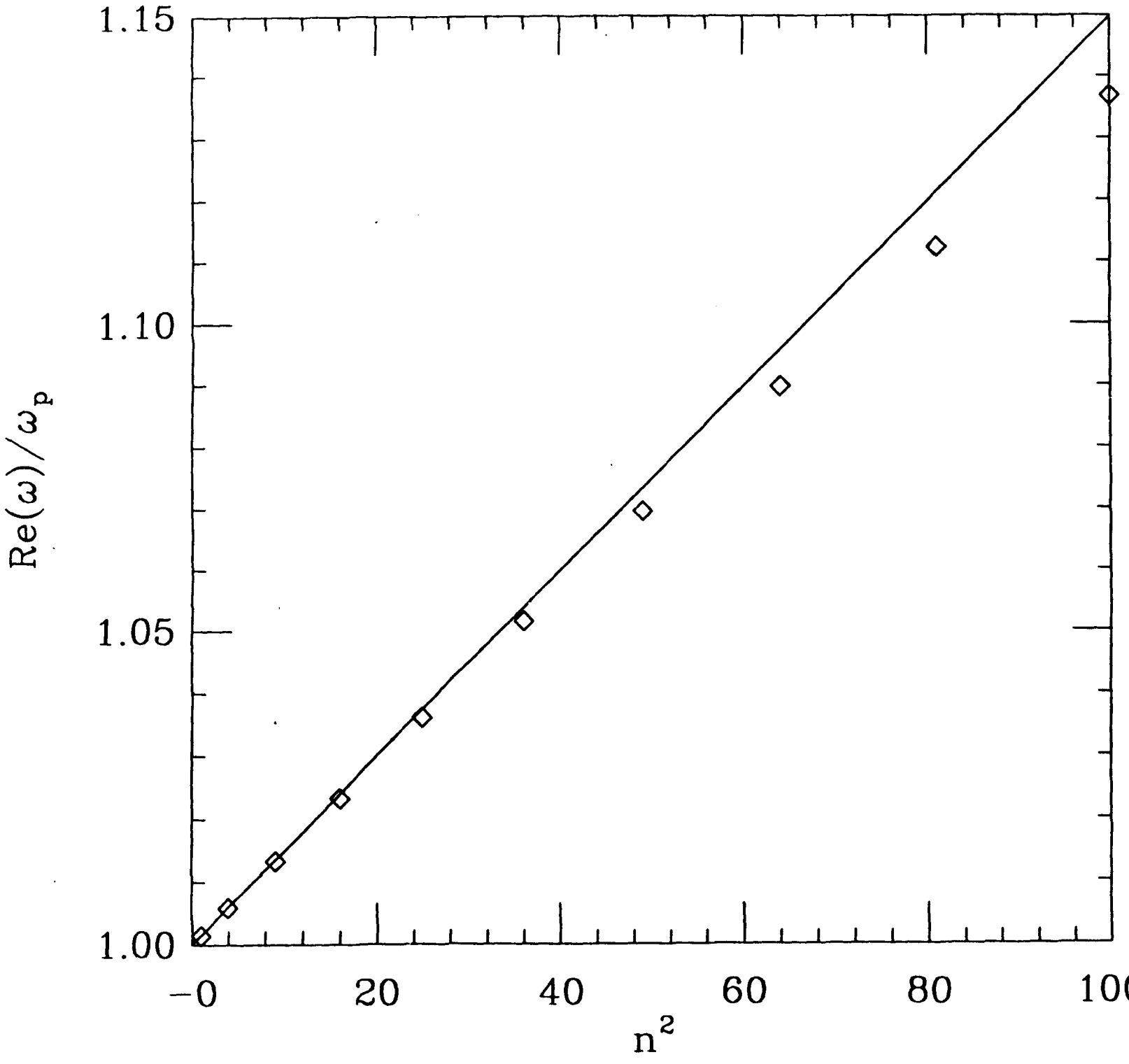

Fig 4a

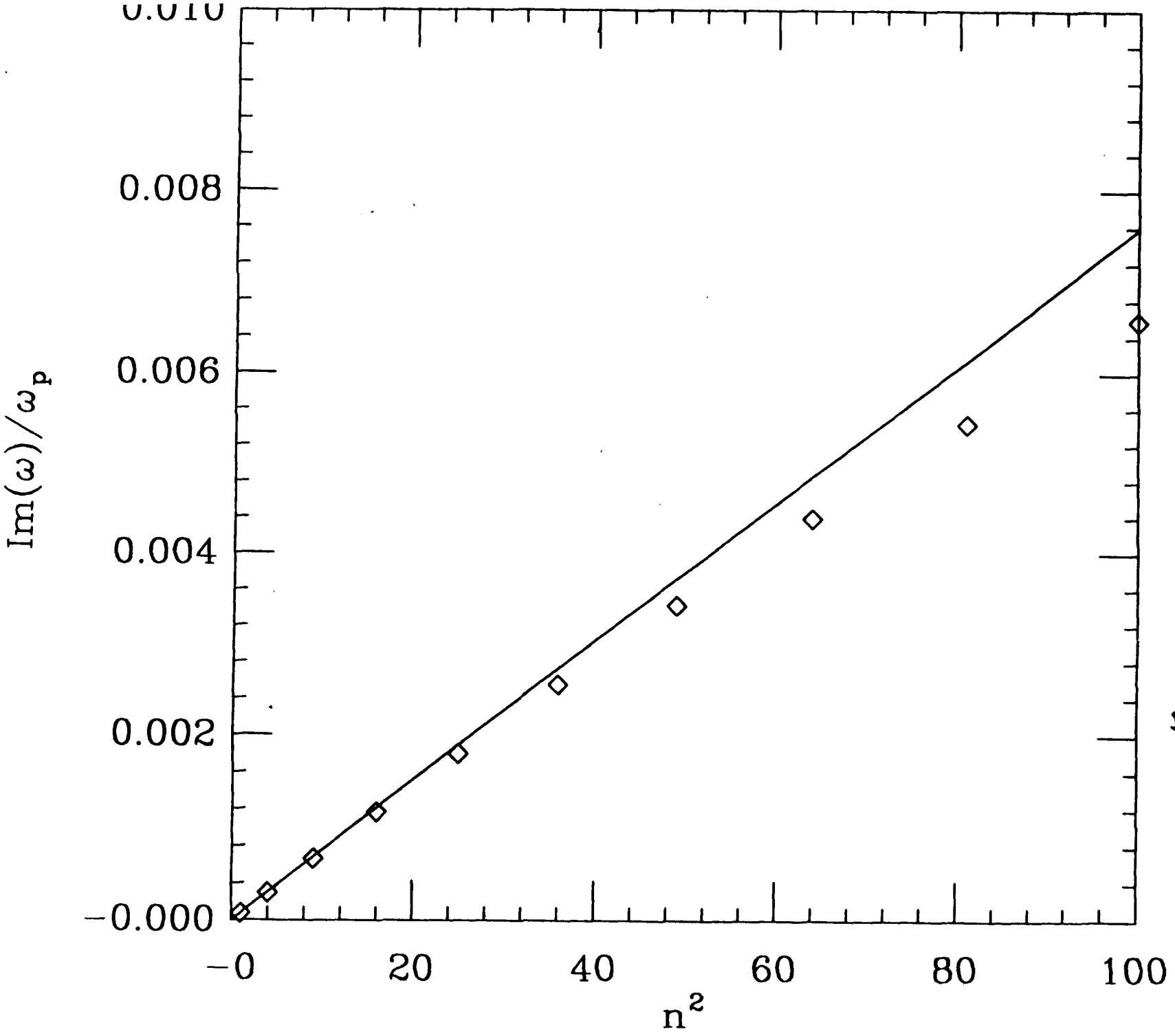

Fig 4b